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Abstract

We have applied thermal insults on LX-04 at 185 ^{o}C and found that the material expanded significantly, resulting in a bulk density reduction of 13%. Subsequent detonation experiments (3 cylinder tests) were conducted on the thermally-damaged LX-04 samples and pristine low-density LX-04 samples and the results showed that the fractions reacted were closed to 1.0. The thermally damaged LX-04 and pristine low-density LX-04 showed detonation velocities of 7.7 to 7.8 mm/μs, significantly lower than that (8.5 mm/μs) of pristine high-density LX-04. Detonation energy densities for the damaged LX-04, low-density pristine LX-04, and hot cylinder shot of LX-04 were 64.8 kJ/cm^3, 66.2 kJ/cm^3, and 65.8 kJ/cm^3, respectively, lower than the detonation energy density of 81.1 kJ/cm^3 for the high density pristine LX-04. The break-out curves for the detonation fronts showed that the damaged LX-04 had longer edge lags than the high density pristine LX-04, indicating that the damaged explosive is less ideal.

Keywords: HMX, LX-04, Thermal Damage, Cylinder Test, Detonation Velocity, Detonation Energy, Edge Lag

1. **Introduction**

Accidents caused by fire, earthquakes, and others (transportation) can expose energetic materials (EM) to unexpected heat and impacts that may damage the explosive charge (e.g., change microstructure, introduce voids and porosity, increase surface area). Mechanical damage and thermal damage are two types of damages that may affect material properties, sensitivity, safety, and performance of energetic materials. Mechanical damage comes from tensile or shear straining, rigorous vibration, or high-rate impact. This may cause deformation of EM and cracking of particles and change its mechanical properties and physical properties. When EM is subjected to unexpected heat, the thermally induced gassing at elevated temperature may result in microcracking, voids, defects, and thermal expansion. It may also cause phase change (e.g., beta to delta transition in HMX).

LX-04 is an HMX-based plastic-bonded explosive, which consists of 85% wt. and 15% wt. binder (Viton A). We have conducted thermal damaged experiments on LX-04 and other HMX-based plastic-bonded explosives and analyzed their properties. Characterization methods and changes in properties (density, sound speed, moduli, gas permeability, porosity) have been reported elsewhere by Hsu et. al [1,2]. After thermal insults, material mass losses were insignificant but volume of damaged samples expanded by more than 10%. Both gas permeabilities and burn rates of the damaged samples increased by several orders of magnitude due to higher porosity and lower density. Moduli of the damaged materials decreased significantly, an indication that the materials became weaker mechanically. Some of damaged samples were also evaluated at room temperature for their sensitivities to impact, friction and spark (small-scale safety tests). Urtiew et. al reported that heated LX-04 was more sensitive to shock initiation at high temperature [3]. Run distance to detonation was shorter for heated LX-04 as the sample was hit by a high-speed impactor.

In this article, we will describe our experimental approach and results in conducting damage experiments on LX-04, performing cylinder tests on damaged samples, and data analysis.

2. Experimental

Before conducting thermal damage experiments, a calculation was done to determine the critical temperature and times to explosion at different temperatures. This enabled us to determine safe heating temperatures and heating durations to avoid unwanted thermal explosion. The information also allowed us to determine a safe operational envelop for thermal damage experimentation. Table 1 shows the times to explosion calculation for cylindrical pressed parts 25.4 x 25.4 mm and 25 x 300 mm, where the latter is the size used in the Cylinder test.

The basic behavior is thermal swelling of LX-04 in an unconfined environment at 185 ^{o}C for two hours in a shot tank. This was done on the 25 x 25 mm pellets of sample 757, and the weight loss was less than 0.1%. The data listed in Table 2 is of two types. Bulk densities were calculated from the dimensions before and after, and these showed a 13% swelling. True densities of samples were measured with a gas pycnometer before and after and this showed a 6% swelling. An X-ray CT scanning technique was used to examine the samples for cracks and voids, and the result is shown in Figure 1.

The cylinder test is a means of measuring energy output of a detonating explosive [4]. Twelve pellets of explosive, nominally 12.7 mm radius (1 inch diameter) by 300 mm long, are placed in a copper tube with both ends open to the air. The copper wall thickness is typically 2.6 mm or about 1/5th of the cylinder radius (called "full-wall"). The explosive is detonated at one end and the shock wave is propagated through the remainder of the material at very high velocity (> 5 mm/μs). The detonation velocity is measured with timing pins installed at several locations along the copper wall. A high-speed streak camera, Fabry-Perot interferometer, or Heterodyne velocimeter is used for recording the radial velocity of the copper wall as it expands following the detonation. From these measurements, the detonation energy and detonation velocity can be measured.

Three Cylinder tests were run and their dimensions are listed in Table 3. Shot 757 was LX-04 previously expanded by heat and the nominal 1-inch size was too small. A larger copper tune was machined to fit the larger pellets. This shot was done at room temperature. Shot 758 were LX-04 pellets ram-pressed at lower pressures to get a density similar to that obtained by the previous thermal expansion. Thus the expansion was put in without heat, and this sample was fired also at room temperature. Shot 759 was pressed to high density at room temperature but was thermally expanded inside the copper cylinder by shining infrared heating lamps. A larger custom copper cylinder was made in anticipation of the swelling. All the larger cylinders were machined to have walls 1/5th the radius. Shot 759 was our first Cylinder shot done at 185^{o}C. The LX-04 cylinder was heated at the temperature for two hours before it was fired. Several IR heating lamps were used as the heating source. Prior to shot 759, a mock assembly with all pins and wires was heated at 185 ^{o}C to ensure that the system could hold up well under the thermal insult.

Figure 2 shows shot 759 in the tank before firing. The cylinder is upright in the center with the detonator at the bottom. The two pin rings are visible that give the detonation velocity far enough down the cylinder that we believe it is steady-state. At the top end, there is a slit over which a glass slide is placed. The emerging detonation front ionizes the air and creates light which is measured by a streak camera, thereby giving a record of the detonation front breakout.

3 Results

The most accurate measured quantity is the detonation velocity, listed in Table 2. It is measured using timing pins, and we list the value near the end of the cylinder where steady state is reached. The detonation velocity depends on material density and can be estimated from the empirical formula shown in Eq. (1).

$$U_S(low) \approx \left(\frac{\rho_{O(low)}}{\rho_{O(high)}} \right)^{2/3} U_S(high) \qquad (1)$$

where U_s is the detonation velocity and ρ_o is the initial bulk explosive density. If we take the average of many dense LX-04 shots as being high, we calculate the corresponding low values expected for less dense LX-04 at the same diameter. The values are fairly close to those measured, so that we believe the similar total energy probably exists in the explosive. For shot 759, the pristine high density LX-04 was thermally damaged at 185 C for 2 hours followed by detonation at temperature. The detonation velocity was measured at 7.820 mm/μs, very close to that for unheated shot 758. The material density for shot 759 right before the shot occurred was estimated at 1.649 g/cc based on the measured detonation velocity. Figure 3 shows a plot of detonation velocity versus density for the two room temperature shots where we know the initial density plus dense LX-04 Cylinder shots going back 40 years.

Is the higher #757 value in Figure 3 above caused by the size (or diameter) effect? We only have two sizes to compare with: 12.7 and 25.4 mm radii (1 and 2 inches). We take only the points that lie between 1.86 and 1.87 g/cc. We get:

1 inch average 8.477 $\pm$ 0.02 mm/μs 3 shots
2 inch average 8.466 $\pm$ 0.04 mm/μs 7 shots.

Using this, we cannot see a size effect at all, and so we cannot attribute this to the difference in our samples. One possible explanation is that, since HMX phase transition occurs at 165°C, for shots 757 and 759, both material might be partially in the δ phase, as opposed to shot 758 which was in β phase [5]. More experiments are needed to draw a more concrete conclusion.

Detonation energy can be determined by the LLNL cylinder test. The cylinder test is a 40 year-old method for measuring the detonation energy at specific relative volumes of expansion, ranging from relative volumes of about 2 to 7 [6]. The essence of the cylinder test is to measure the wall velocity and equate the square of this velocity to the detonation energy. Because the detonation is confined, a good estimate can be made of the average relative volume in the space behind where the wall measurement is

being made. In other words, we can imagine the cylinder to be like a balloon, where internal pressure pushes out a volume, which can be calculated. The first approach was to equate the square of the copper wall velocity with energies of explosives believed to be near-ideal [7]. Then, the calculated detonation energies from the thermo-chemical code CHEETAH became good enough to be used as the standards for full burn [8]. The three standard relative volumes were reset to 2.2, 4.4 and 7.2 for scaled wall displacements of 6, 12.5 and 19 mm, respectively. For scaling, a 12.7 mm radius (1 inch diameter) tube was taken as the standard. In scaling, all data is reduced to this standard size. At LLNL, the streak camera was replaced by Fabry-Perot interferometry and now by Ted Strand's Hetereodyne Velocimetry [9]. The two laser methods are more accurate than the streak camera, and the Heterodyne is cheaper than the Fabry. The error bars for streak camera detonation energies are set at $\pm$3%, although much of this may be unknown material differences. The error bars for the laser methods are unknown because nothing has been repeated, but $\pm$1% is the probable lower limit.

The time output of our shots is shown with older data in Figure 4. Our data are the lower curves, which represent the actual measurement with Heterodyne velocimetry. The upper curves that are 1 inch full-wall, high density pristine LX-04 (#88 and 120/21 combined) are polynomial fits to the streak camera data and contain extensive smoothing. One 2-inch, half-wall shot has been scaled (divide time, displacement by 2; divide velocity by $2^{1/2}$) and the original data is shown, with the entire scatter inherent in the streak camera.

The measured wall velocities go into a Gurney-type equation, which keeps the density of the copper wall constant with expansion [10]. The equation is

$$F = \frac{E_d}{E_d(Ch)} = \frac{\alpha\beta\rho_o}{E_d(Ch)}\left[\frac{\rho_m}{\rho_o}\left(\frac{R+x}{R_o}\right)^2 ln\left(\frac{R+x}{R}\right) + \frac{1}{4}\left(\frac{R+x}{R}\right)^2\right]u_m^2 \qquad (2)$$

The result is the burn fraction, F, at some given relative volume, which is described with the measured wall velocity u_m. R_o is the initial radius, R the radius at time t and x the wall thickness at time t. The initial densities are ρ_m for the metal and ρ_o for the explosive. E_d is the calculated detonation energy and E_d(Ch) the detonation energy from CHEETAH. The coefficients α and β are experimental adjustments for the method of measurement, as described above, and the wall type. This equation allows us to successfully calculate early full-wall shots that used metals other than copper and to adjust for small variations in dimensions.

The detailed results of the cylinder tests are given in Table 3. All LLNL results are shown, with ambient LX-04 tests going back over 40 years. The resulting detonation energy, E_d, is then compared with

that calculated by the thermo-chemical code CHEETAH and the fraction reacted, F, at any relative volume is given by

$$F = \frac{E_d(measured)}{E_d(CHEETAH)} \tag{3}$$

The slightly larger size of 757 and 759 is included in the scaling. Shot #757 was 1.05 times larger than 12.7 mm radius, so that time and displacement (but not velocity) are divided by 1.05 to get the scaled results. The resulting detonation energies may be used to make an Equation-of-State. The burn fractions are averaged across the three volumes. We see that LX-04 never detonates 100% in the Cylinder test but the expanded material actually seems slightly more efficient.

We plan to conduct higher degree of thermal damage (higher temperature and confinement) on LX-04 and perform more cylinder tests in order to gather more information on detonation velocity, detonation energy, and equation of state data for damaged LX-04.

The measured break-out curves are shown in Figure 5. Our new tests #757 #758, and #759 are off-axis, which easily happens with explosive parts, which are imperfect and more difficult to line up. It does not detract from the important issue, which is the size of the edge lag, i.e. the displacement at each edge. These are listed in Table 4. The average edge lag is a rough measure of the reaction zone length. The edge lags for HMX and high density pristine LX-04 are smaller than those for the expanded material, which shows that the latter is less ideal. The edge lag for low density pristine LX-04 gets larger because the explosive needs more volume to collect energy from to drive the detonation front.

Summary

We applied thermal insults on LX-04 at 185 ^{o}C and found that the material expanded significantly, resulting in a bulk density reduction of 13%. Subsequent detonation experiments (3 cylinder tests) were conducted on the thermally-damaged LX-04 samples and a pristine low-density LX-04 samples. The LX-04 samples were heated at 185 ^{o}C for two hours in an unconfined environment and allowed to cool down to room temperature for characterization and the first cylinder test. The second cylinder test was performed on a pristine LX-04 whose density was identical to that of the first case. A third sample was pressed to high density and was heated in place inside the copper cylinder at 185^{o}C for two hours, which was then fired at 185^{o}C. The thermally damaged LX-04 and pristine low-density showed detonation velocities of 7.7 to 7.8 mm/μs, significantly lower than that (8.5 mm/μs) of pristine high-density LX-04. Detonation energy

densities measured at 7.2 relative volume (19.0 mm scaled distance) for the damaged LX-04, low-density pristine LX-04, and hot cylinder shot of LX-04 were 64.8 kJ/cm^3, 66.2 kJ/cm^3, and 65.8 kJ/cm^3, respectively, lower than the detonation energy density of 81.1 kJ/cm^3 for the high density pristine LX-04. The resulting detonation energy density, was then compared with that calculated by the thermo-chemical code CHEETAH. The fractions reacted (F) were found to be 0.98, 1.0, and 0.97 at the relative reactive volume of 7.2 for the damaged LX-04, low-density pristine LX-04, and hot cylinder shot of LX-04, respectively. The break-out curves for the detonation fronts showed that these three cylinder tests had longer edge lags (0.95 mm, 0.90 mm, and 1.25 mm, respectively) than the high density pristine LX-04 (0.60 mm). These larger edge lags showed that the damaged LX-04 is less ideal explosive than the high density pristine LX-04.

In summary, LX-04 was able to withstand a thermal event at 185 ^{o}C for 2 hours and still retained its energy, albeit spread out in a larger volume which resulted in lower detonation velocity and detonation energy density.

Acknowledgments

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Table 1. Times-to-explosion calculations for cylindrical pressed parts of LX-04.

Temp.	Time (hours)	
(^{o}C)	25 x 25 mm	25 x 300 mm
210	1.8	2.1
200	3.5	3.4
190	7.0	6.6
180	16	16.2

Table 2. Swelling data for three Cylinder shots and thermal swelling**.**

Shot Number	757	758	759	Historical
Swelling Method	heated	pressed low	heated	none
Cylinder Temp.	ambient	ambient	hot	ambient
Initial Bulk Density (g/cm^3)	1.864	1.867	1.84	1.864-1.869
Final Bulk Density (g/cm3)	1.638	1.636	1.649 est.	
% Bulk Swelling	-12	-12	-10	
Initial True Density (g/cm^3)	1.867			
Final True Density (g/cm3)	1.759			
% True Swelling	-6			
Explosive Diameter (mm)	26.4	25.37	26.1	
Explosive Length (mm)	322.4	309.4	304.8	
Copper Tube Length (mm)	323.6	304.8	323.7	
Detonation Vel. (mm/μs)	7.801	7.697	7.820	8.49-8.51
Calc. Det. Vel. (mm/μs)	7.784	7.795		

Table 3. Cylinder test results for all LLNL LX-04 shots.

Shot No.	Wall Matl	Explosive Radius (mm)	Wall Thick (mm)	Measured Scaled Wall Velocity (mm/μs) 6	12.5	19	Detonation Energy, E_d (kJ/cm^3) 2.2	4.4	7.2	Burn Fract.
Historical- full density										
73	Al	25.41	14.64	1.340	1.568	1.644	5.74	7.86	8.64	0.96
88	Cu	12.71	2.60	1.513	1.646	1.701	6.30	7.45	7.96	0.94
90	St	12.71	2.61	1.540	1.668	1.720	6.53	7.66	8.14	0.97
156	Th	12.71	2.04	1.492	1.663	1.726	6.02	7.48	8.05	0.94
437	Cu	25.41	2.72	2.072	2.239	2.331	6.46	7.54	8.17	0.96
547	Cu	25.43	2.71	2.062	2.245	2.310	6.38	7.56	8.01	0.95
547	Cu	25.43	2.71	2.002	2.158	2.231	6.29	7.31	7.81	0.93
This report-expanded										
757	Cu	13.27	2.59	1.369	1.491	1.555	5.02	5.96	6.48	0.97
758	Cu	12.72	2.55	1.360	1.491	1.551	5.09	6.12	6.62	0.99
759	Cu	13.16	2.56	1.399	1.544	1.603	5.34	6.23	6.58	0.98

Table 4. Measured average edge lags from detonation front curvature in 25 mm-radius LX-04 copper cylinders.

Cylinder test number	Sample type	Edge lag (mm)
597	High density pristine	0.6
598	High density pristine	0.6
757	Damaged	0.95
758	Low density pristine	0.9
759	Hot cylinder test	1.25

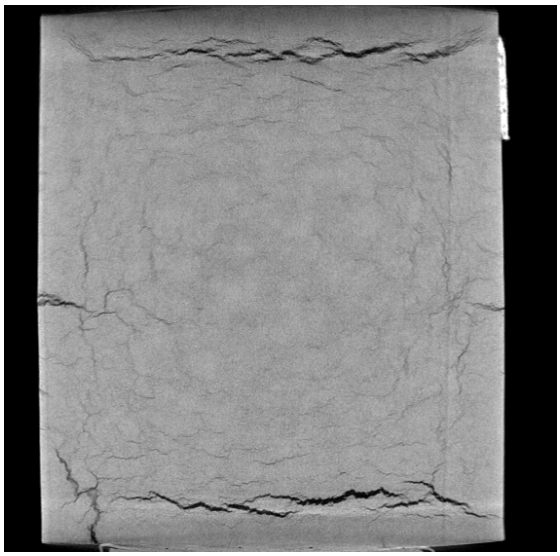

Figure 1. X-ray CT pictures of damaged LX-04 from side view of the cylindrical part.

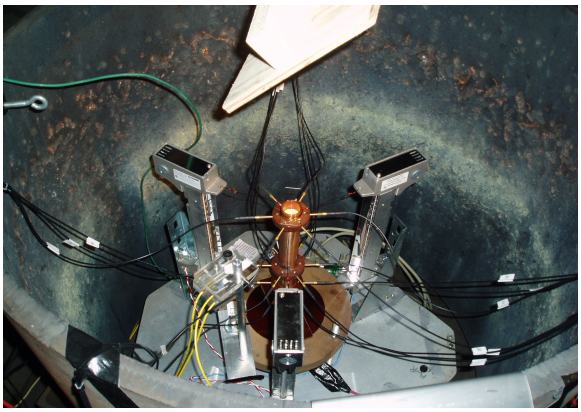

Figure 2. Hot cylinder test #759 in a secondary containment inside a 1.0-kg shot tank.

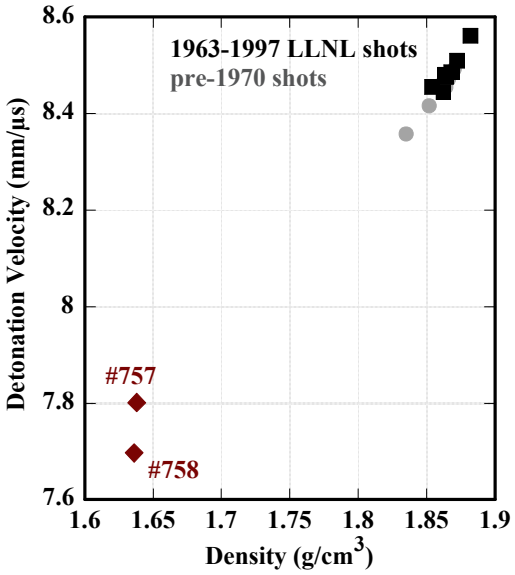

Figure 3. Detonation velocity of LX-04/copper cylinders as a function of density.

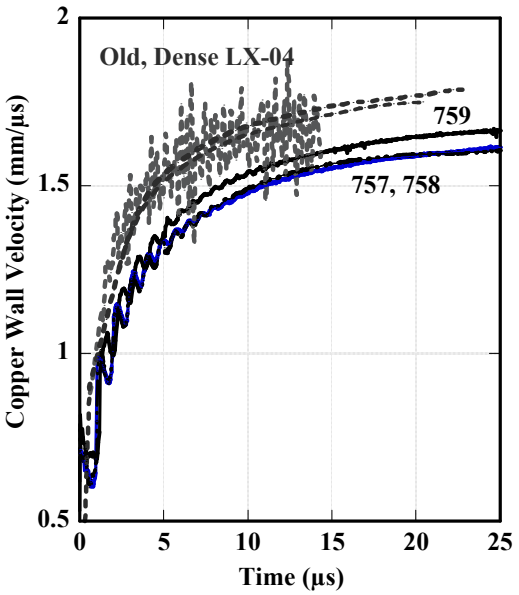

Figure 4. Comparison of new data with two old full-wall 1-inch shots (gray dashed 88, 120/121). A 2-inch half-wall shot (scattered gray 437) has been scaled and shows the scatter inherent in the streak camera. The swollen LX-04 shots lie below with 759 at the top and 757 and 758 overlying each other below.

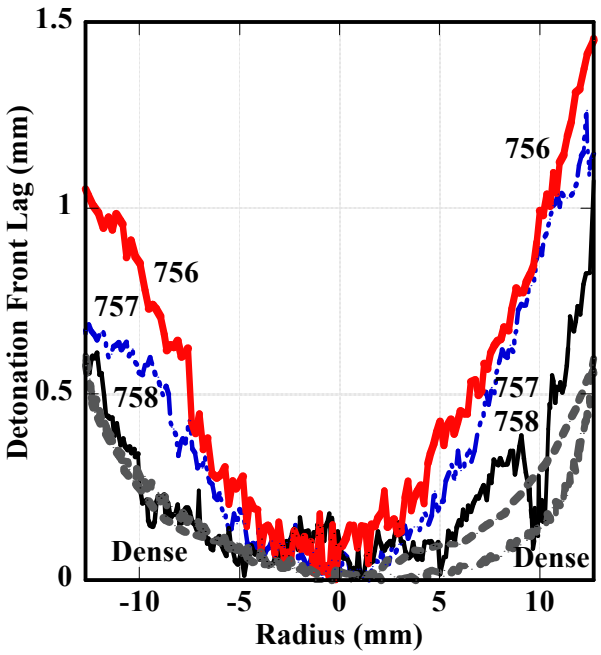

Figure 5. Detonation front breakout curves for the new data (757, 758, 759) and two fully dense LX-04 copper cylinders (597, 598). The edge lags are larger with the less dense explosive.